

Large Area SrTiO₃ Thin Films for Active Microwave Applications Grown by Pulsed Laser Deposition

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Abstract—Thin films of SrTiO₃ have been grown by pulsed laser deposition (PLD) on 2" diameter LaAlO₃ substrates using a technique in which the laser beam is rastered over the radius of a 4 inch diameter target while the substrate is rotated in a radiative blackbody heater. The deposition occurs through a pie shaped opening in the heater. The structure, composition and thickness have been measured using XRD and RBS. The dielectric properties of the film have been measured at 1 MHz utilizing an LCR meter. The results indicate that the films are single phase and (001) oriented. However, film composition, thickness and dielectric properties are not uniform along the radius of the film. The results indicate that the composition nonuniformity is due to off-axis deposition, and correlates with changes in the dielectric properties.

I. INTRODUCTION

Ferroelectric materials exhibit an electric field dependent dielectric constant. Recent reports of the electric field effect in bulk and thin films suggests that these materials could be used to develop active microwave electronics such as phase shifters, tunable filters and tunable, high Q resonators [1,2,3]. Ferroelectrics are chemically and structurally compatible with high temperature superconductors (HTS). Devices based on HTS and ferroelectrics offer a significant improvement over normal metal ferroelectric structures. In fact, the combination of ferroelectrics and superconductors for these applications has recently been patented [4]. SrTiO₃ is a candidate material for low temperature applications because of its low losses at microwave frequencies in single crystal form ($\tan\delta \ll 10^{-3}$) and its compatibility with HTS. Although bulk SrTiO₃ is not ferroelectric it still has a field dependent dielectric constant. Since the electric field required to achieve a maximum change in the dielectric constant is on the order of 100 kV/cm, thin film applications are desirable because these fields can be achieved with bias voltages of ≤ 10 V.

For many applications, such as multi-pole band pass and band reject filters operating in the cellular frequency range (800 MHz to 2 GHz), large area films ($> 1"$) are required. In

order to fabricate a balanced multi-pole filter, the film properties must be uniform across the entire area of the film. The properties of most importance are the field dependence of the dielectric constant, loss tangent and film thickness. We have deposited SrTiO₃ thin films by pulsed laser deposition (PLD) on 2" LaAlO₃ substrates. The structure, composition, thickness and dielectric properties of a representative thin film have been investigated across the surface of the film to characterize the uniformity over large areas.

II. EXPERIMENTAL

Thin films of SrTiO₃ were grown on 2" diameter (001) LaAlO₃ by PLD. The PLD system used to grow these films has been described in more detail elsewhere [5]. A KrF excimer laser (~ 30 nsec pulses, ~ 300 mJ/pulse and $\lambda = 248$ nm focused with a 50 cm focal length lens to a fluence of ~ 2 J/cm²) was used to ablate a 4" diameter target of SrTiO₃

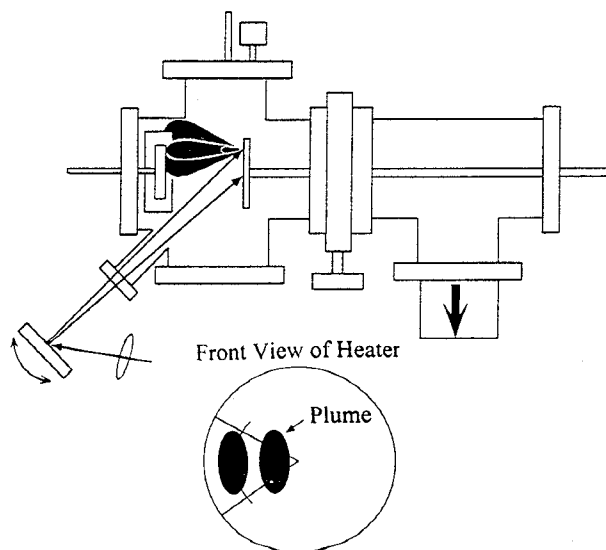


Fig. 1. Large area pulsed laser deposition chamber.

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obtained from Coating and Crystal Technologies. The vaporized material was deposited onto a radiatively heated 2" LaAlO₃ substrate positioned approximately 11.5 cm away from the target as shown in Figure 1. The substrate was heated to 725°C in an oxygen ambient pressure of 300 mTorr. The films were deposited at approximately 0.092 Å/pulse (2.3 Å/sec @ 25 Hz) to a total film thickness of ~1300 Å. The films were then cooled to room temperature in oxygen at ~10°C/min. A photograph of one of the films is shown in Figure 2. The films are optically transparent and very smooth. A 1 cm² reference film was grown in another chamber. The differences in the growth conditions are the small raster area, a 3.5 cm target-substrate distance and a conductive heater with no shielding.

Structural measurements were made by x-ray diffraction using a Rigaku Rotaflex diffractometer with Cu K_α radiation from a rotating anode source. Rutherford Backscattering Spectroscopy data was collected using ⁴He⁺ at 5.560 MeV at six spots, ~3 mm increments, along a radius of the film to determine compositional homogeneity and thickness. Half of the wafer was cut into 15 mm square pieces. Two pieces along the radius were used to measure the dielectric properties near the center and the edge. Measurements of the dielectric properties were made using silver interdigital electrodes (~1.5 μm thick) on the surface of the films. A matrix of Ag electrodes with varying dimensions were patterned by a multilayer photoresist process. The dielectric measurements reported here were all made on 6 pole electrodes with a gap spacing of 5 μm, a finger width of 7.5 μm and a finger length of 75 μm. Electrical contact was made by wire bonding gold wires to the large contact pads. The test fixture utilized a four point probe approach. The capacitance was measured as a function of temperature (30 - 300 K) and with dc bias voltages (0 - 40 V in 5 V increments, 0 - 80 kV/cm) at 1 MHz using an HP4285A LCR meter. The temperature



Fig. 2 Picture of a SrTiO₃ thin film grown on a 2" LaAlO₃ substrate with a reflection of the Naval Seal.

control was achieved with an APD Cryogenics closed-cycle refrigerator with a Lakeshore 330 temperature controller. A background measurement was made on a LaAlO₃ substrate without a film which showed a negligible temperature and dc bias field dependence. Thus changes in the capacitance as a function of temperature are due to changes in the dielectric susceptibility of the film.

III. RESULTS

A. Structural Measurements

All of the SrTiO₃ films were found, by x-ray diffraction, to be single phase and well oriented. Figure 3 shows a $\theta/2\theta$ scan for the SrTiO₃ thin film grown on the 2" LaAlO₃ wafer. The x-ray beam exposed an area of the film along the film radius thus the x-ray scan shows a structural average of the film. The narrow ω - scan, shown in the inset of Figure 3, is near the resolution limit of the diffractometer using 0.16° slits, indicating that the crystal growth out of the substrate plane is extremely well oriented. Previously we have analyzed smaller films at the National Synchrotron Light Source which indicated that the films were nearly single crystal in structure [6].

B. Composition and Thickness

Figure 4 shows the RBS spectra at six positions along the radius of the 2" film. The positions are indicated in the inset of Figure 4. The data was fitted using RUMP simulations with the background due to the substrate subtracted out. From the fitting we were able to determine the compositional and thickness variations across the film. The Sr/Ti ratio in the film is an average 15% below the target composition and there is a systematic increase in the Sr/Ti ratio from the edge to the center which exceeds the error of the experiment, ~±5%. From Figure 4 the average thickness has been found

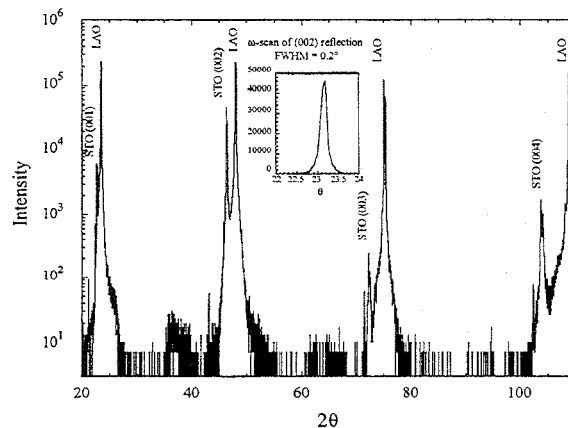


Fig. 3. X-ray diffraction pattern of a SrTiO₃ thin film grown on a 2" LaAlO₃ substrate. Inset shows an ω -scan for the (002) reflection in the film.

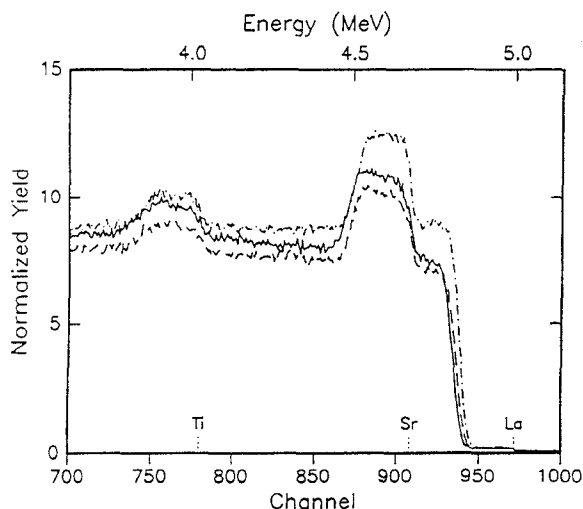


Fig. 4. RBS Spectra acquired at several positions along the radius of a SrTiO_3 thin film grown on a $2''$ LaAlO_3 substrate.

to be approximately 1300 Å. The change in thickness is approximately 15% between the center and the edge, where the edge is the thickest.

C. Dielectric Properties

The dielectric properties were measured at the center and the edge of the SrTiO_3 film deposited on the $2''$ wafer as well as the reference film. Two 1 cm squares were cut from the $2''$ wafer along a radius so that dielectric measurements could be made near the center and edge of the film. The measurements were made at 1 MHz as a function of temperature while cooling for several dc bias electric fields.

In Figure 5, the capacitance is presented for the center and edge pieces of the large area film, as well as, for the reference film. All the films were measured with dc bias voltages applied between 0 and 40 V (≤ 80 kV/cm across a 5 μm gap). The basic temperature dependent features of the

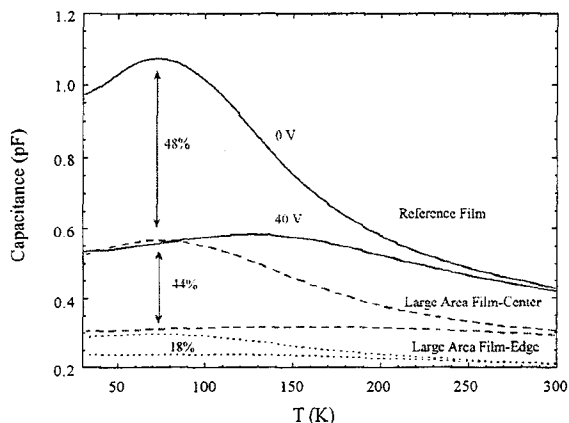


Fig. 5. Capacitance as a function of temperature with a dc bias.

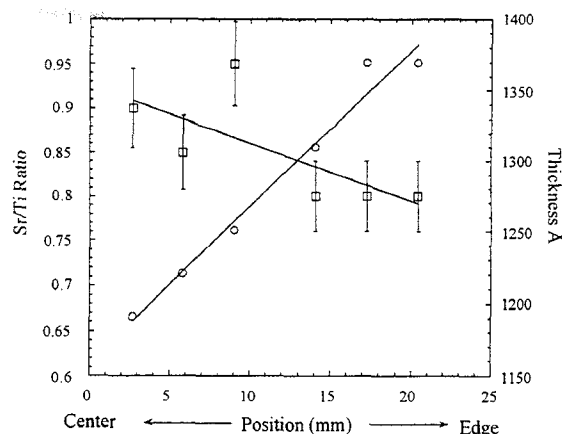


Fig. 6. Composition and thickness of a SrTiO_3 thin film grown on a $2''$ LaAlO_3 substrate as determined by RBS at several positions along the radius of the film.

capacitance for this material have been described previously [7]. The main difference between these three films lies in the sensitivity of the film to the applied dc bias field. The tunability (percent change in the capacitance under an external dc bias) is reduced in the large area film as shown in Figure 5. The maximum tunability of the reference film is 48% where as the maximum tunability in the large area film is as much as 44% at the center and drops off to 18% at the edge.

IV. DISCUSSION

The loss of tunability and the lack of uniformity in the dielectric properties is critical to the use of SrTiO_3 thin films for device applications. The RBS results indicate a nonuniform thickness and a non-stoichiometric Sr/Ti ratio which is also nonuniform across the radius of the film.

The RBS analysis in Figure 6 shows the thickness variation from the center to the edge to be $\sim 15\%$ where the center of the film is thinner than the edge. Typically thicker films have better dielectric properties, in particular better tuning [8]. For this large area film, the opposite is true. This indicates the non-stoichiometry to be the likely cause for the reduced dielectric tuning at the edge of the film. It has been shown in YBCO that off-axis material does not have the same composition as the on-axis material [9]. Early films of SrTiO_3 grown over large areas also gave evidence that off-axis material was not the same as on axis material. The first film grown in the chamber used a very slow raster rate for the laser. X-ray analysis of this film indicated a superlattice structure in which the periodicity matched the period of the laser raster. This would indicate that there is some difference between the on-axis material and the off-axis material.

Figure 6 also shows the Sr/Ti ratio for several positions between the center and the edge of the film. The data indicate that the Sr/Ti ratio is off-stoichiometry by 15% on average, and there is a systematic change in the composition between the center and the edge of the film. This initially seems

counter intuitive since the center of the film is constantly exposed to the deposition. The most probable cause for improved stoichiometry at the center of the film results from the heater design. Figure 1 shows a front view of the heater and substrate. Deposition occurs through a 60° wedge shaped opening in the black body heater. When the plume rasters to the edge of the film the substrate is exposed to full on-axis and off-axis deposition, but when the plume is rastered toward the center, the covering of the heater blocks much of the off-axis material. This results in an improved stoichiometry at the center of the film and therefore improved dielectric properties.

V. CONCLUSIONS

We have grown SrTiO₃ on a 2 inch LaAlO₃ substrate by PLD. The film was analyzed by XRD, RBS, and dielectric measurements. Structurally, the film is single phase and well oriented, but RBS indicated some nonuniformity in both thickness and composition. The average film thickness is 1300 Å which increased linearly from the center to the edge by 15%. This could have been improved simply by using a progressive rate raster for the laser beam to compensate for the change in translation speed of the substrate along its radius. The Sr/Ti ratio is 15% low on average and varies along the radius of the film decreasing from the center to the edge. This compositional deficiency correlates with reduced dielectric tuning and is attributed to compositional variations in the plume. The off-axis region of the plume is expected to

be off stoichiometry. The simplest solution to this would be to mask off the off-axis material. This will result in a reduced deposition rate, but presumably improved dielectric properties.

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